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Magnetic properties of (Ga,Mn)As digital ferromagnetic heterostructures

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Magnetic properties of (Ga,Mn)As digital ferromagnetic heterostructures have been investigated by polarized neutron reflectometry and magnetometry. T_c of three samples with 20, 50, and 100 ML GaAs spacers ranges from 30 to 40 K. The saturation magnetization of three samples exhibits a pronounced tail extending over 50 K above T_c in addition to a temperature-independent background. For the 50 ML sample, PNR measurements show a similar tail but no background. These behaviors can be explained by a two-step ordering process. In the tail region, two-dimensional islands first individually become ferromagnetic. Long-range order develops as the temperature is decreased below T_c. © *2004 American Institute of Physics.*
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III–V-based ferromagnetic semiconductors, such as (Ga, Mn)As in the form of superlattices or random alloys, are a class of materials that has received much attention in recent years due to their interesting physical properties and the potential for spintronic device applications.[1–5] The present understanding of the magnetism in these systems derives mainly from experiments based on bulk magnetization measurements and magneto-transport measurements, both of which probe the average magnetization over the whole sample. These techniques have proven quite useful for study of the random alloy. However, in layered structures such as digital ferromagnetic heterostructures (DFH), magnetic ordering occurs in quasi-two-dimensional sheets. In addition, the interaction among these well-defined magnetic layers may lead to a T_c enhancement. Polarized neutron reflectometry (PNR) is well known[6] for its unique ability to probe such layered magnetic structures and provide spatially sensitive information on the magnetic ordering and interlayer interactions.

Here we present a study of the magnetic properties of (Ga,Mn)As DFHs using PNR. The DFH samples were grown by low-temperature molecular beam epitaxy (MBE) on semi-insulating GaAs (100) substrates and the details have been reported elsewhere.[3] In the three DFHs studied, each bilayer unit consists of a 0.5 ML MnAs and a GaAs spacer layer of varying thickness t ($t=20$, 50 and 100 ML). The number of bilayer repeats N is 60, 150 and 100. We denote the DFH structures by $(t,0.5)_N$. For comparison, we have also studied two $Ga_{1-x}Mn_xAs$ random alloy samples with $x=0.055$ and 0.080. Superconducting quantum interference device-based magnetization measurements show that the T_c ranges from 30 to 40 K for the DFH samples and 60 to 70 K for the random alloys.

The PNR measurements were carried out on the NG-1 reflectometer at the NIST Center for Neutron Research using neutrons of wavelength $\lambda=4.75$ Å. In our experiments, the neutrons were polarized by Fe/Si supermirrors positioned before and after the sample, with an efficiency >97%. With the use of two Al-coil spin flippers, we measured all four polarization cross sections, $R^{(+,+)}$, $R^{(-,-)}$, $R^{(+,-)}$ and $R^{(-,+)}$ as a function of the wave vector Q,[7] where $Q=4\pi\sin(\theta)/\lambda$ and θ is the angle of incidence (and reflection) of the specularly scattered neutrons. The "+" and "−" signs denote the parallel and antiparallel neutron polarizations, respectively, of the incident and reflected neutrons with respect to a guide field applied in the sample plane. The nonspin-flip (NSF) scattering cross sections $R^{(+,+)}$ and $R^{(-,-)}$ are sensitive to both the chemical (scattering from the nuclei) and magnetic ordering, whereas the spin-flip (SF) scattering cross-sections $R^{(+,-)}$ and $R^{(-,+)}$ are purely of magnetic origin. Specifically, the difference between the two NSF cross sections provides information about the in-plane

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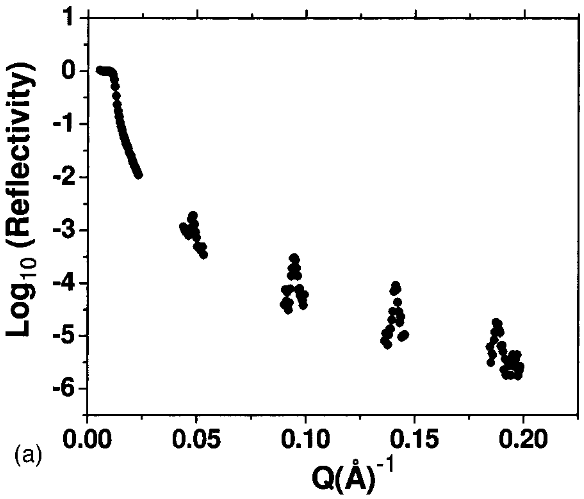

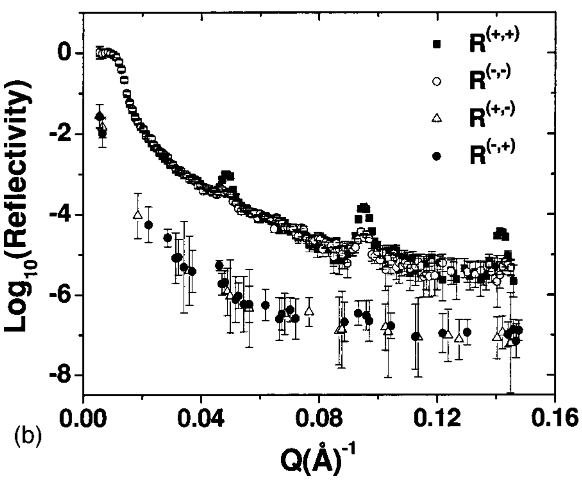

FIG. 1. (a) Unpolarized neutron reflectivity for the $(50,0.5)_{60}$ DFH at room temperature, showing good structural and interface quality of the superlattice. (b) Polarized neutron reflectivity for $(50,0.5)_{60}$ DFH measured in a 0.9 Oe field along the easy axis after cooling in zero field to 15 K. These data have been corrected for the polarized efficiencies of the polarizing elements.

magnetization component parallel to the guide field while the two SF scattering cross sections are sensitive to the in-plane magnetization component perpendicular to the guide field.[7]

Figure 1(a) shows an unpolarized neutron reflectivity scan of a DFH sample $(50,0.5)_{60}$. For small Q, total internal reflection of neutrons occurs and the NSF reflectivity equals 1. Above the critical Q, the neutrons transmit through the sample and the reflectivity decreases. The four regularly spaced peaks are the Bragg peaks up to the fourth order of the superlattice (SL) structure. The presence of these well-defined SL peaks is an indication of the good structural quality of the sample. The spacing between the adjacent peaks is related to the average bilayer thickness d, by $Q=2\pi/d$. The bilayer thickness is found to be about 14 nm, in good agreement with the nominal thickness of the structure.

A typical PNR scan for the same sample is shown in Fig. 1(b) after cooling in nearly zero field (<0.15 Oe) to 15 K. At low temperature, a small guide field of 0.9 Oe was applied in the sample plane along a [110] in-plane cleave axis of GaAs. The SL peaks in the NSF scattering are split, while the SF scattering is negligible after correcting for the efficiencies of the polarizing elements. The splitting of the NSF cross sections at the SL peaks indicates that the magnetization has a component along the polarization direction of the incoming neutrons, whereas the absence of any significant SF scattering indicates that there is negligible magnetization perpendicular to the polarization direction of the incoming neutrons. Even though the DFH sample was cooled in nearly zero field, the Mn layer moments are ferromagnetically aligned parallel to the small guide field, which is much smaller than the coercivity (~ 30 Oe) of the sample at 15 K, as determined from the magnetometry measurements. It is possible that the sample exhibits spontaneous ferromagnetic interlayer coupling upon cooling through T_c, as proposed by Kepa[8] for GaMnAs/GaAs superlattices. To test this idea we performed a series of PNR and bulk magnetization measurements after cooling in small positive and negative fields ($|H|<0.2$ Oe) applied along the easy axis. The results suggest that only a small residual field is required to set the layer magnetization direction through the entire DFH sample as it is cooled through T_c (where the anisotropy is arbitrarily small).

To explore further the behavior of the DFH samples near T_c, we measured both the spontaneous magnetization M_r and saturation magnetization M_s of the DFH samples in the magnetometer as a function of temperature in a constant field applied along the easy [110] direction. M_r and M_s are rather different [Fig. 2(a)]. First, M_r is very small for $T>T_c$ ≈ 38 K, while M_s is very large and appears to be offset vertically from M_r. This background is temperature independent and persists even to room temperature. We speculate that this background may originate from small ferromagnetic Mn particles or clusters with a very high T_c, as discussed below. After the constant background in M_s is removed, both M_r and M_s are shown in the inset of Fig. 2(a). Obvious in the inset is the long tail in M_s that extends well above T_c. The width of this tail is about 60 K, greater than T_c itself. In addition, M_s is greater than M_r through the entire temperature range indicating an excess moment aligned along the external field direction. A similar phenomenon was also observed in two other DFH samples. Another unusual feature in M_s is the steep slope of the magnetization at low temperatures, which is different from that in M_r. For comparison, M_s for the random alloy samples does not show either a pronounced tail or a steep slope at low temperatures [Fig. 2(b)].

To understand the origin of the excess magnetic moment evident above T_c, we probed the $(50,0.5)_{60}$ DFH sample using PNR in fields applied parallel to the [110] easy axis and measured the temperature dependence of the NSF splitting at the first SL peak [Fig. 1(b)]. (Note that the difference in the integrated intensity $\Delta R=R^{(+,+)}-R^{(-,-)}$ at this peak is proportional to the moment component parallel to the applied field direction.) Figure 3 summarizes the results of these measurements in fields of 0.9 and 20 Oe after cooling in zero field and in a field of 2000 Oe (i.e., saturation). Consistent with the magnetometer results (Fig. 2), the saturation magnetization is larger than the low-field magnetization (in 0.9 and 20 Oe), in particular above $T_c\approx 40$ K. The downturn of the 0.9 Oe data at low temperatures is accompanied by a very slight increase in the SF scattering at the first SL peak

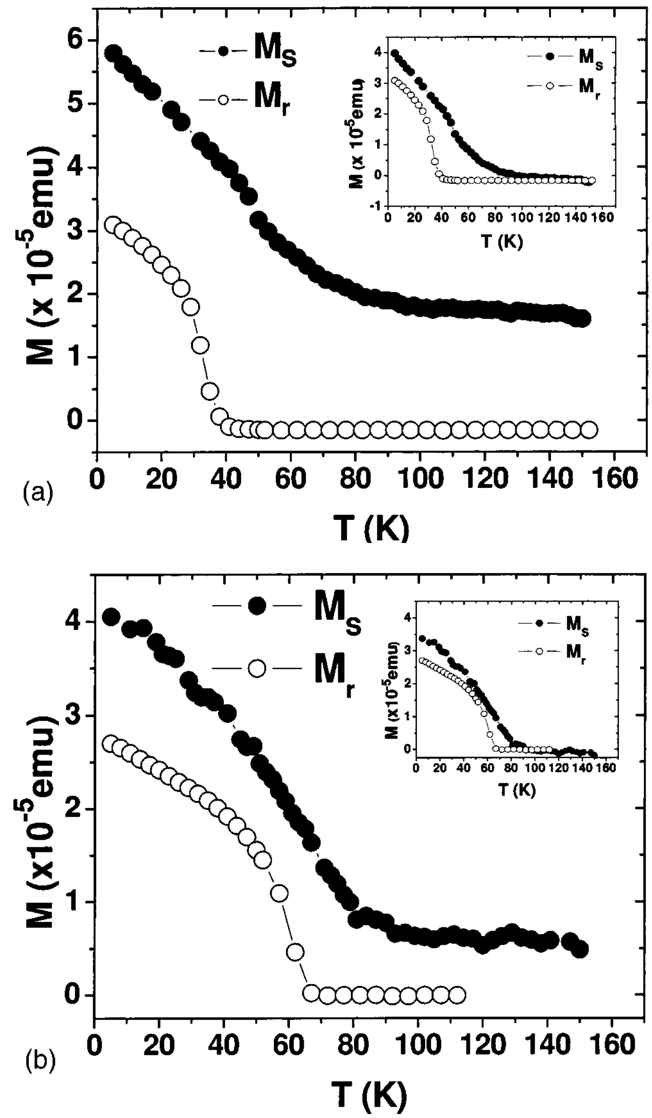

FIG. 2. (a) The temperature dependence of saturation magnetization (M_s) measured in a field of 5000 Oe and spontaneous magnetization M_r) measured in a field of ~0 Oe for the $(50,0.5)_{60}$ DFH. The inset shows the magnetizations after background subtraction. (b) The temperature dependence of saturation magnetization (M_s) in field of 5000 Oe and the spontaneous magnetization (M_r) in a field of ~0 Oe for a 5.5% random alloy. The inset shows the magnetizations after background subtraction.

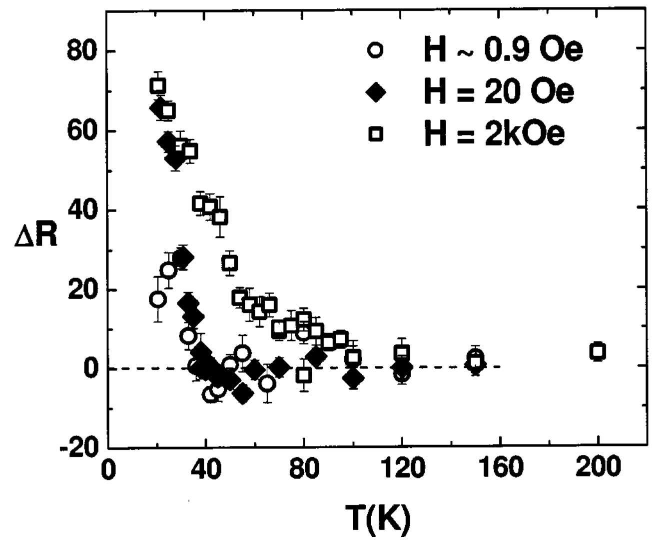

FIG. 3. The temperature dependence of magnetization for the $(50,0.5)_{60}$ DFH determined by PNR. The vertical axis represents the integrated intensity difference $\Delta R = R^{(+,+)} - R^{(-,-)}$ between the NSF cross-sections at the 1'st SL (Bragg) peak position.

position. We conclude that a small component or fraction of the Mn moment is spontaneously oriented parallel to the hard axis upon cooling in zero field, but a 20 Oe field is sufficient to align it.

For the 2000 Oe PNR data, a tail persists above 40 K, but there is no measurable constant high-temperature background (above 100 K) as seen in the magnetometer M_s data. Note that the splitting at the SL peaks in the PNR measurements is sensitive *only* to the magnetization contribution that has the corresponding periodicity of the SL (i.e., Mn moments correlated from one MnAs layer to the next). On the contrary, the magnetometer measurements average over both periodic and random magnetic moments present in the sample. Thus the high-temperature (i.e., above 100 K) background in the magnetometer data can be attributed to *randomly* distributed magnetic clusters lacking the SL periodicity. A field of ± 2 kOe was sufficient to set the temperature independent cluster moment in different directions at $T > T_c$ in separate magnetometer runs. Subsequent measurements of the magnetic hysteresis in small fields at $T < T_c$ showed that the loops were essentially identical except for an offset in the magnetization due to the cluster moment. This suggests that the cluster moments are not coupled with the layer magnetization. The overall features in the PNR measurements, such as the tail evident in saturation above 40 K, agree well with those in the magnetometer data after the high-temperature background is removed [Fig. 2(a) inset]. Clearly, above T_c the Mn moments within the MnAs planes are not correlated from one MnAs layer to the next in small applied fields.

In summary, we have identified two types of contributions to the magnetization of DFHs using PNR and magnetometry. One type has the periodicity of the superlattice, and the other type originates from random clusters. At and below 100 K, magnetic moments within each MnAs layer first order locally and then develop long-range order along the growth direction as the temperature is lowered through T_c. This two-step ordering mechanism may originate from the discontinuous nature of the quasi-two-dimensional MnAs layers.

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